

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
28 December 2000 (28.12.2000)

PCT

(10) International Publication Number
WO 00/79623 A1

(51) International Patent Classification⁷: **H01M 4/96**,
8/08, 8/10

(21) International Application Number: PCT/US00/40268

(22) International Filing Date: 21 June 2000 (21.06.2000)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
60/140,246 21 June 1999 (21.06.1999) US

(81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:

- With international search report.
- Before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments.

(71) Applicant (*for all designated States except US*): **PRO-CYON POWER SYSTEMS, INC.** [US/US]; Suite 138, Building 20, 2701 Monarch Street, Alameda, CA 94501 (US).

(72) Inventor; and

(75) Inventor/Applicant (*for US only*): **MANIKOWSKI, Ambrose, F.** [US/US]; 1179 Stafford Drive, Cupertino, CA 95014 (US).

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(74) Agents: **WHEELLOCK, E., Thomas et al.**; Morrison & Foerster LLP, 755 Page Mill Road, Palo Alto, CA 94304-1018 (US).

(54) Title: **PEAK POWER FUEL CELL**

(57) Abstract: This discusses a new fuel cell technology, capable of providing bursts of high power electrical pulses from a few seconds to ten or more seconds. One advantage of the fuel cell is that it does not need noble metal catalysts to achieve high power densities. Both activation and concentration polarization can be practically eliminated in the pulse fuel cell. The design is based upon the principle that a cathode can be charged with activated oxygen taken from the air. This activated oxygen is adsorbed on carbon (which is a poor electrochemical catalyst). The electrode is then discharged when electrons are added to the activated adsorbed oxygen to form hydroxide ions. The overall electrochemical reaction that takes place is the same as in a conventional fuel cell, with the distinctive difference being that a two step charge/discharge cycle is used to produce a pulse of electrical power. A similar process can be used on the anode to adsorb hydrogen on a metal hydride forming alloy or metal. The fuel cell can be designed to achieve any pulse duration, limited only by the carbon loading on the cathode, and the metal hydride loading of the anode. The oxygen adsorption material for the cathode preferably is activated carbon mixed with graphite along with an inert binder. The anode material is preferably a hydride-forming alloy or metal.

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PEAK POWER FUEL CELL

FIELD OF THE INVENTION

This invention relates to an electrical fuel cell and to its components, specifically to its oxygen-storing cathode. The inventive fuel cell is capable of providing bursts of high power electrical pulses with a duration of a few seconds to ten or more seconds.

BACKGROUND OF THE INVENTION

The concept of producing high-energy pulses of electrical power from fuel cells was investigated in the mid to late 1960's by companies performing research for U.S.

Department of Defense-related programs. Apparently, the research was never carried past the exploratory stage. The following paper describes some of these early efforts: "Pulsed Power Fuel Cells" by Sanderson, Bushnell, McKiernan, of Pratt & Whitney. This paper appeared in Fuel Cell Systems – II, 5th Biennial Fuel Cell Symposium sponsored by the Division of Fuel Cell Chemistry, at the 154th Meeting of the American Chemical Society, Chicago, Ill. September 12 – 14, 1967, Bernard S. Baker, Symposium Chairman.

Other procedures have been selected in the past to store significant amounts of electrical power for delivery over a short period of time. For instance, ultra-capacitors represent a similar technology where electrical energy is stored based on the principle of separation of charged species in an electrolytic double layer. All electrode / electrolyte interfaces exhibit double-layer capacitance; however, only devices that do not exhibit Faradaic reactions are considered Electrochemical Double Layer (ECDL) Energy Storage Devices. See, "The Aerocapacitor: An Electrochemical Double-Layer Energy-Storage Device", by Mayer, Pelka and Kaschmitter of Livermore National Laboratory, Livermore Calif. 1993.

There have been discussions in the literature recently suggesting a hybrid ultra-capacitor and battery. Some researchers ascribe the attributes of both batteries and capacitors to certain devices that are said to exhibit pseudo-capacitance or reversible Faradaic surface reactions. These devices are called ultra-capacitors, and use both capacitance and Faradaic reactions for storage of electrical energy. The ultra-capacitor combines the principles of the electrochemical double layer with bipolar electrodes using a non-aqueous solid state electrolyte for the Faradaic reactions.

The pulse fuel cell exhibits both capacitance and Faradaic reactions taking place between the hydrogen at the anode and the oxygen at the cathode. This is the distinguishing feature of the pulse fuel cell; the Faradaic reactions taking place in the pulse fuel cell are between hydrogen and oxygen. Ultra-capacitors involve Faradaic reactions with other chemical species such as lithium and zinc.

The pulse fuel cell is also distinguished by the feature that the oxygen for the reaction is supplied from air passing through the system. This oxygen is adsorbed and stored on the surface of the active element of the cathode until it is consumed during the power pulse.

The capacitance of the mercury/electrolyte interface is the most thoroughly studied phenomena related to ECDL's. From these studies, theoretical capacitance values of 10-20 micro-Farads/cm² are obtained for single electrodes in concentrated aqueous electrolytes. The charge stored in a capacitor charged to 1.2 volts is given by the formula

$$Q = CV$$

and the energy stored is

$$E = 1/2 CV^2$$

where C is the capacitance of the device, and V is the voltage to which it is charged. The theoretical charge and energy stored in a capacitor at 20 micro-Farads/cm² charged to 1.2 volts is 14.4 micro Joules/cm².

Recently, ultra-capacitors using high surface area electrodes composed of activated carbon composites or RuO₂ based metal oxide electrodes have been studied. Activated carbon and activated carbon fiber cloths can be produced inexpensively with up to 2000 m²/gram of surface area. This value would correspond to a single theoretical electrode capacitance of 400 Farads/Gram, if all the surface area were electrochemically accessible. With an application of 0.060 g of carbon/cm², a capacitance of 240 Farads/cm² results. Charged to 1.2 volts, this would lead to storage of 288 coulombs/cm², or 172 Joules/cm² of stored energy.

The above theoretical electrode capacitance of 400 Farads/gram has not been achieved to date because the complete surface area is not accessible. Typical activated-

carbon ultra capacitors store about 40 Farads/gram, or about 1/10th of the theoretical value, leading to charge storage of 28.8 coulombs/cm² and energy storage of 17.2 joules/cm².

Lawrence Livermore National Laboratory has developed an carbon aerogel with a carbon density of 0.5 grams/cm³, and a specific capacitance of 35 Farads/gram of carbon capable of storing 25 coulombs/cm². Other investigators claim to have demonstrated (100 Farads/cm²), or 75 coulombs/cm², approaching about 25% of the theoretical limit of 288 coulombs/cm².

In contrast, we have demonstrated using our inventive pulse fuel cell technology (a combination of graphite/activated carbon) to produce an electrode surface that has good electrical as well as oxygen adsorption properties. This electrode was 0.3 mm thick, and was comprised of 0.035 grams of carbon/cm², about an order of magnitude less carbon than used in the ultra capacitors, with a surface area of 1150 m²/gram. The charge storage approached 1 coulomb/cm² in electrodes containing 30 mg carbon/cm².

In pulse fuel cell tests using the inventive devices, current densities in excess of 1.0 Amp/cm² are seen which suggests that the theoretical pulse length of 22 seconds are possible, based upon a charge storage of 22 coulombs/cm². Tests to date achieved 1/10th of the theoretical value, or 2.2 coulombs/cm² for a pulse duration would be 2.2 seconds.

These ultra-capacitors operate on a much shorter time scale than the pulse fuel cell, typically less than 10 milliseconds for full discharge. The pulse fuel cell current density measurements that have been made to date are typically at 0.3 to 3 seconds.

Even though other technologies are possibly capable of the desired storage capacity, the inventive fuel cell is much more able to deliver energy in a lengthier, controlled pulse as is desired in automotive usage.

Commercialization of fuel cells requires significant reductions in cost. The first low temperature fuel cells used high noble metal catalyst loading of 4 to 10 mg/cm². Today the lowest catalyst loading reported are on the order of 1 mg/cm². This invention will significantly reduce or completely eliminate use of noble metal catalysts in fuel cells.

The inventive power fuel cell may use a conventional (high performance) hydrogen electrode at the anode or an anode comprising a metal hydride forming alloy. A carbon-rich cathode, preferably with little or no noble metal catalyst, may be used to adsorb oxygen from the air. During the pulse discharge period, the adsorbed hydrogen and oxygen are consumed to produce water and electrical power in the conventional manner.

The inventive pulse fuel cell system may be used in a variety of applications. For instance, it can be designed to produce a high current pulse to power an automobile during acceleration and hill climb, the most demanding phase of automobile operation. The power pulse for this application may be on the order of 40 kWe for 10 to 15 seconds during vehicle acceleration, and 20 kWe for 5 minutes for extended hill climbing. In another application of the inventive fuel cell, less demanding pulse discharges of less than a second are useful for fuel cell applications such as distributed electrical power generation and co-generation, where economics is the primary concern, and lowest capital cost of the fuel cell is desired.

The inventive pulse power fuel cell differs from a conventional fuel cell in three important ways:

- 1.) Activated carbon stores a quantity of pure oxygen from air required for the power pulse within the cathode of the fuel cell.
- 2.) The fuel cell operates at ambient temperature, and requires no noble metal catalyst, only Carbon/Graphite are used in the cathode.
- 3.) A train of short electrical pulses is produced where each pulse is 0.1 to 10 seconds in duration depending on the application.

Activated carbon stores a quantity of pure oxygen from air required for the power pulse within the cathode of the fuel cell. The fuel cell operates at ambient temperature, and requires no noble metal catalyst, only carbon/graphite are used in the cathode. A train of electrical pulses is produced where each pulse is 1 to 5 seconds in duration for a period of 10 to 20 seconds (or sufficiently long for the oxygen to be replenished at the cathode for the next pulse). This high power pulse train may be input to an electrical motor controller to produce traction power.

Procyon's inventive pulse fuel cell concept essentially eliminates the concentration and activation polarizations, the major internal electrochemical loss in fuel cells operating at high power. The length of the high current output pulse from the fuel cell is limited only by the quantity of oxygen adsorbed on the carbon, and the quantity of oxygen dissolved in the electrolyte. For example, for the most demanding automotive PULSE power requirement, approximately 45 grams. of oxygen is adsorbed on activated carbon in the fuel cell cathode will meet the coulomb charge balance requirements of a 40 kWe pulse for 10 seconds. By taking advantage of oxygen that is stored on the carbon at the electrode, this invention both improves voltage output and extends the pulse width of power from a fuel cell. When all adsorbed oxygen has been consumed, the fuel cell returns to its normal

performance on air where the concentration polarization associated with air limits output current. Figure 1 illustrates the increased power output of a conventional PEM fuel cell under steady-state conditions represented by sweep rates of 0.1 to 3 seconds. High power pulse output associated with short sweep rates of a fraction of a second.

5 Most fuel cells are capable of high current density, high power operation for a short time period. The duration of this pulse of power is limited by quantity of oxygen adsorbed on the carbon within the cathode of the fuel cell. Figure 1 depicts an example of these phenomena as measured using a conventional PEM fuel cell with platinum catalyst. The current-voltage output is shown at dynamic sweep rates from 0.10 second (the pulse mode)
10 to 10 seconds, the steady state output. Approximately 30% increase in power at 0.5 volts is noted for relatively short pulse duration of less than 0.1 seconds. The short pulse period is limited by the low carbon loading which is about 1 mg/cm^2 for this PEM (proton exchange fuel cell).

The duration of pulse that can be obtained from conventional fuel cells is generally
15 limited to less than 1 second, because of the limited quantity of carbon used in the cathode. A fuel cell can be designed to provide a longer duration pulse if the quantity of oxygen adsorbed or otherwise stored near the electrode reaction site can be increased. Pulse power can be obtained from alkaline (KOH) fuel cells, acid (Phosphoric) fuel cells as well as PEM (proton exchange fuel cells as exemplified by Figure 1). It is possible to adapt the pulse
20 power concept to PEM fuel cells for modest pulse duration's on the order of seconds. However, this inventive pulse power alkaline fuel cell development is capable of producing power pulses up to 10 seconds in duration. This is because the high carbon loading required to demonstrate 10 second pulses can more easily be implemented with alkaline technology. Conventional alkaline fuel cells typically contain $20 \text{ mg Carbon /cm}^2$, as
25 compared to 1 mg/cm^2 in a typical PEM.

Of course, there are other known fuel cells.

Figure 2 shows the inventive pulse power fuel cell using an alkaline fuel cell with KOH electrolyte and compares it to a conventional alkaline fuel cell to illustrate the concept.

30 These processes result in nearly eliminating two important electrochemical losses in a fuel cell: a) the concentration polarization, and b) the activation polarization. A description of these two polarizations and their effect on fuel cell performance is described in Figures 3 and 4. The length of the high current output pulse from the fuel cell is limited

primarily by the quantity of oxygen adsorbed on the carbon, at the cathode. Some fuel cell types may limit pulse duration through other mechanisms. For instance, in alkaline fuel cells, the quantity of oxygen dissolved in the electrolyte forming OH⁻ ions may also limit pulse duration.

5 Assuming that the power pulse in a hybrid automotive power system should be on the order of 40 kWe for 10 seconds during vehicle acceleration, approximately 40 grams of pure oxygen adsorbed on activated carbon in the fuel cell cathode will meet the coulomb charge requirements. Referring to Figure 3 where a cell current density of 1000 ma/cm², at 0.5 volts will require 200 cells of 400 cm² area to produce 40 kWe power at 100 volts and
10 400 amps. This reduces to a requirement to store 0.50 mg O₂/cm² on the cathode. Assuming 0.50 grams of O₂ is adsorbed on 1 grams of carbon (see Figure 6) then 0.50 mg of O₂/cm² is equivalent to 10 mg of Carbon/cm². Tests to date have achieved 2.5 seconds pulse duration on 44 mg Carbon/cm².

 All fuel cells are capable of high current density, high power operation for a short
15 time period. The duration of this pulse of power is limited by quantity of oxygen adsorbed on the carbon within the cathode of the fuel cell. None of the cited documents nor any of the known prior art describes a fuel cell of the type or performance described herein.

BRIEF DESCRIPTION OF THE DRAWINGS

20 Figure 1 shows the pulse output of a PEM fuel cell.

 Figure 2 is a depiction of the inventive pulse power fuel cell compared to atypical alkaline fuel cell.

 Figure 3 is a graph showing polarization curves of conventional fuel cell compared with the inventive peak power fuel cell.

25 Figure 4 is a graph depicting polarization curves of a conventional fuel cell vs. the inventive peak power fuel cell.

 Figure 5 is a graph depicting OH⁻ ion concentration between electrodes as a function of time.

 Figure 6 is a graph depicting the amount of oxygen adsorbed on activated carbon as
30 a function of pressure.

 Figure 7 is a graph depicting the discharge of several fuel cells (including an inventive fuel cell) at a constant current discharge.

Figure 8 is a graph depicting the performance of silver and carbon cathodes (including an inventive cathode) with oxygen

Figure 9 is a graph depicting the performance of the inventive carbon cathodes with oxygen.

5 Figure 10 is a graph depicting hydrogen electrode polarization as a function of temperature.

Figure 11 depicts the results of various methods for extending pulse width.

Figure 12 shows a configuration for various multiple electrodes.

10

DESCRIPTION OF THE INVENTION

This inventive fuel cell and constituent cathode is intended to supply high current density, high power operation for a comparatively longer time than is expected from prior art devices. The duration of this pulse of power is enhanced by the quantity of oxygen
15 adsorbed on the carbon within the cathode of the fuel cell.

The duration of this pulse from conventional fuel cells is generally limited to less than 1 second. My fuel cell provides a longer duration pulse because the quantity of oxygen adsorbed or otherwise stored near the electrode reaction site is increased. Pulse power can be obtained from either alkaline (KOH) fuel cells, and possibly from phosphoric
20 acid fuel cells, or from polymer electrolyte membrane (PEM) fuel cells using the concepts of this invention.

My approach to the pulse power fuel cell results in the following benefits:

1) The concentration polarization and activation polarization are nearly eliminated in the pulse power fuel cell, thereby enabling greatly increased current density compared to
25 conventional steady state fuel cell operation.

2) Sufficient activated carbon, at least 75 mg/cm^2 , along with a metallic current collector such as silver or nickel comprise the fuel cell cathode which is capable of storing sufficient oxygen 100 mg/cm^2 to meet automotive peak power demands of 10 seconds or more

30 3) Circulating sufficient air/oxygen through the fuel cell allows easy generation of a 10 second pulse. The pulse may consist of a sequence of shorter pulses (referred to as a "pulse train") where each individual pulse may be on the order of fractions of a second to many seconds, depending upon the amount of oxygen-adsorbent materials (and their

adsorbance capacity) integrated into the cathode. A number of pulses may be produced to generate the necessary power over the 10-second time interval.

4) The preferred embodiment combines activated carbon and graphite to obtain optimum properties of oxygen adsorption from activated carbon, and electrical conductivity from graphite.

5) Carbon and graphite are typically the only active elements at the cathode; no precious metal catalyst is required on the cathode electrode, and the pulse fuel cell operates best at ambient temperatures where oxygen adsorption is maximized. Included in the desired carbon material suitable for use in the inventive cathode and fuel cell are the carbon aerogel products discussed above. The carbon loading within the cell and the electrical conductivity using these aerogels will both be increased.

6) Alternate materials may be used to enhance the oxygen storage of the pulse power technique. These alternate materials, such as Ag/AgO, Ni(OH)₂ and Pt(OH) are capable of a weak reaction with OH⁻ ions, where oxygen is stored in the form of hydroxide. These OH⁻ ions can be released by small voltage swings, typically 0.5 volts in the case of Ni(OH)₂.

The inventive pulse power fuel cell is based upon the principle of operation that oxygen adsorbed on the active carbon elements of the fuel cell provides the oxidant to the electrochemical reaction during the transient period where pulse power can be obtained. A conventional fuel cell depends upon diffusion of oxygen to the active electrochemical reaction site. This diffusion process creates a concentration polarization formed by the transport of oxygen through inert nitrogen in air, and through the electrolyte/gas interface. This concentration polarization is the major energy loss mechanism in a conventional diffusion-limited fuel cell. This is the most significant and unique aspect of pulse operation of a fuel cell. The pulse power fuel cell is capable of high current density, high power operation for the duration of the electrical pulse. The duration of the pulse of electricity is limited by the amount of oxygen stored on the activated carbon in the fuel cell cathode, and the availability of a OH⁻ ions stored in the electrolyte on the surface of the hydrogen electrode. In the limit, at t=0, the fuel cell has a polarization curve that is limited by the hydrogen 1/2 cell. This high current density is only available at the beginning of the pulse, and exponentially decays to the polarization curve shown for air (see Figure 3) which typically takes less than 1 second for a conventional fuel cell. To date we have shown in

our tests that this decay time can be extended from 0.69 second for carbon loading of 20 mg Carbon /cm², to 2.2 seconds for 44 mg/cm². The average power that can be drawn from the pulse fuel cell operated over multiple on/off cycles will be similar to that of a conventional fuel cell performance with air.

5 The activation concentration is another major energy loss in a conventional fuel cell. Our tests have shown that the activation concentration is also practically eliminated during pulse operation. As seen in Figure 4, the capacitance effect increases the voltage output near open circuit voltage for the fast polarizations (25 amps/sec). For the slower polarizations (2.5 A/sec and 0.25 A/sec) a linear reduction of voltage with current density is
10 noted, which indicates a resistive voltage drop. A non-linear drop in voltage (about 0.05 volts) near open circuit characterizes the activation polarization in a conventional fuel cell, and transition to a linear voltage drop with current, which is due to internal resistance of the cell. As seen in Figure 4, there is no evidence of activation polarization in either the fast sweep (25 A/sec) nor in the slow sweep (0.25 A/sec) representing the steady state
15 performance of a conventional fuel cell.

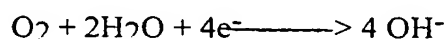
It is also noted with reference to Figure 4, that the fast polarization curves for air and oxygen are nearly identical, while the slow polarizations are significantly different for air and oxygen, which is characteristic of conventional fuel cell performance.

20 The oxygen stored within the fuel cell is available for supplying instantaneous peak power. Three sources of oxygen storage are available:

- a) Oxygen adsorbed on the carbon at the cathode, (Faradaic effect)
- b) Oxygen gas contained within the flow passages of the fuel cell
- c) Oxygen stored as OH⁻ in the electrolyte of the fuel cell, (psuedo-capacitive effect) for alkaline fuel cells.

25 To demonstrate the functional characteristics of the pulse fuel cell, estimates have been made of the peak current and the time duration of this current pulse for a conventional fuel cell. These computations are compared to the test data obtained on the inventive carbon/graphite cathode. Estimates have also made of the quantity of carbon required to design a cathode to produce a pulse of ten seconds duration. It is assumed that the fuel cell
30 is operated at average current of 1.0 Amps/cm² during the pulse. The following equations used to estimate the pulse time and the width of the depletion layer are taken from the "Handbook of Fuel Cell Technology" Prentice Hall, 1968, Dr. Carl Berger, Editor. The inventive carbon/graphite fuel cell cathode is used as the model for these calculations.

An electrode containing a quantity of electrolyte sufficient to provide a high power pulse output from a fuel cell is described in Figure 5. The OH⁻ stored in a typically 5 molar solution of KOH contains approximately 6.9×10^{-3} mole equivalents of OH⁻ per cubic centimeter of solution. Oxygen in air going into solution with water at the surface of the oxygen electrode typically has a concentration of 8.0×10^{-6} moles per cubic centimeter at atmospheric pressure. Each mole of oxygen produces 4 moles of OH⁻ according to the following:



Drawing current from the fuel cell will deplete the OH⁻ ion concentration at the hydrogen electrode until the concentration of OH⁻ is reduced to nearly zero. At the time that the OH⁻ ion concentration at the hydrogen electrode becomes zero, steady state operation is achieved, and the current generated by the fuel cell becomes diffusion limited by the rate that the OH⁻ ions can diffuse through the electrolyte according to the following equation:

$$J_L = nFDC_b/\Delta$$

The assumed value of J_L , the fuel cell discharge current, is 1 amp/cm² when operating at steady state with oxygen from air. From this equation the depth of the diffusion layer Δ is determined by:

$$\Delta = nFDC_b/J_L$$

The diffusion coefficient D for OH⁻ ions in KOH electrolyte is estimated to be about 1.5×10^{-5} cm²/sec¹, $F = 96,500$ coulombs/gram, and C_b (the concentration of 30% KOH) is 6.9×10^{-3} moles/cm³ equivalent, and $n = 1$ electron participating in the electron transfer. The diffusion length Δ , when steady state current is achieved is;

$$\Delta = 1(96500)(1.5 \times 10^{-5})(6.9 \times 10^{-3})/1.0 = 0.010 \text{ cm.}$$

The total number of electrons drawn during the transient where the concentration of OH^- ion in the diffusion layer is reduced to zero at the surface of the hydrogen electrode is:

$$\begin{aligned} (\Delta Q) &= nF\Delta C_b/2 \\ 5 \quad &= 1(96500)(0.0285)(6.9e^{-3})/2 \\ &= 3.32 \text{ coulombs/cm}^2 \end{aligned}$$

Again, this compares favorably with the coulomb storage we have measured on the inventive carbon/graphite cathodes (30 mg Carbon/ cm^2), which was 3.2 coulombs/ cm^2 at the slow sweep rate of 0.25 Amps/sec, and 0.935 coulombs/ cm^2 at the high sweep rate of 25 Amps/sec. This 3.32 coulombs/ cm^2 is the total charge that is adsorbed on the carbon. At the high sweep rates (25 Amps/sec), only a fraction of this total stored charge (0.29 coulombs/ cm^2) is utilized. Considering a transient current of 1 Amp/ cm^2 , then this surface charge of 3.32 coulombs/ cm^2 can support a pulse of this magnitude for:

$$15 \quad T = \frac{3.32 \text{ coulombs/cm}^2}{1.0 \text{ Amps/cm}^2} = 3.32 \text{ sec}$$

Once the total stored charge is depleted, the fuel cell current output becomes limited by the diffusion of OH^- ions through the electrolyte. The maximum current that can be drawn from the fuel cell is the limiting current J_L and steady state operation that is shown in Figure 3 is 200 mA/ cm^2 for the air polarization curve.

During the pulse, the output voltage drops from open circuit voltage to about 0.5 volts, and the current rises to the concentration polarization free current which is shown in Figure 3 to be 1000 mA/ cm^2 . The voltage and current will remain at this high level until the OH^- ions in the electrolyte reservoir become depleted, or the oxygen available at the cathode is consumed.

To generate a second power pulse, oxygen at the cathode of the fuel cell must be replenished from the air. For alkaline fuel cells, the OH^- ion reservoir adjacent to the hydrogen electrode must also be recharged with OH^- ions generated at the cathode. By opening the circuit between the fuel cell and the load and permitting the voltage in the fuel

cell to recover to its open circuit value of 1.0 volt, the balance of the OH⁻ ions in the electrolyte adjacent to the hydrogen electrode is restored.

Figure 6 indicates the oxygen adsorption on activated carbon. This Figure shows that for sugar carbons, and 1 atmosphere pressure, 0.050 grams of oxygen are adsorbed per gram of carbon. The carbon/graphite fuel cell cathodes we have used contain 0.035 grams of carbon per cm², and is theoretically capable of adsorbing 0.00175 grams of oxygen per cm². The transition time for a 1.0 amp/cm² current density is:

$$T = M_{O_2} n F / J_L, \quad T = (0.00175/32)(4)(96500)/1.0, \quad T = 21.1 \text{ seconds}$$

The theoretical value of charge stored is:

$$\Delta Q = n F M_{O_2} / (\text{molecular wt}) \text{Area}_{\text{cell}} = 4(96500)(0.00175)/32/1.0 = 21.1 \text{ coulombs/cm}^2$$

Our test data using a constant voltage discharge at 0.50 volts, and 0.375 amps/cm² we have measured pulse durations of up to 2.56 seconds, which results in charge storage of 1.0 coulombs/cm² about 1/20 of the theoretical value of 22 coulombs/cm² as calculated above..

The following table summarizes the previous theoretical computations that were made in estimating the potential of a fuel cell to deliver pulses of electrical power, with experimental data reported herein.

	Theoretical	Measured at 0.375 A/cm ²
Electrolyte	3.32seconds	1.60 seconds
O ₂ adsorbed on C	<u>21.1 seconds</u>	<u>1.00 seconds</u>
Total	24.42 seconds	2.60 seconds

The value shown for the electrolyte is only applicable to alkaline fuel cells. The oxygen available from the air in the fuel cell passages is quite small and will replenish only a very slight amount of the depleted oxygen in the fuel cell. In PEM fuel cells (and for alkaline fuel cells that are not limited by OH⁻ ions in the electrolyte) the ultimate limit on

the pulse duration from the pulse power fuel cell is the amount of oxygen adsorbed on the activated carbon at the cathode.

The above range of transition times is based upon diffusion coefficients for OH⁻ ions in KOH of 1.5×10^{-5} cm²/sec, which gives this value for diffusion of OH⁻ ions in KOH.

5 The above calculated range of values for transition times compares well with the transition times associated with our tests of the inventive carbon / graphite electrodes, which ranged from 0.69 to 2.56 seconds.

Although all fuel cells are capable of high current density, high power operation for a short time period. The capacitive effect produced by movement of ions between the
10 electrodes, and the Faradaic effect associated with the quantity of oxygen adsorbed on the carbon within the cathode limits pulse duration. My fuel cell provides a longer duration pulse because the quantity of oxygen adsorbed or otherwise stored in the carbon near the electrode reaction site may be increased by either more effective utilization of the carbon, or by increasing the carbon loading on the cathode.

15 Another advantage of the inventive pulse fuel cell is the small amount of heat generated at high power levels. The pulse fuel cell can be operated at high power levels and at higher efficiency than conventional fuel cells because the concentration and activation losses are extremely low. The major thermal loss in this fuel cell is internal resistance of the cell. The pulse fuel cell operates on a relatively low duty cycle where very
20 high power levels are generated for short time periods (i.e. possibly as long as 22 seconds if all carbon is fully utilized (as seen in the above table). followed by longer periods of quiescent operation. When used in an automotive drive cycle, the quiescent period of the fuel cell is very long compared to the pulse power period. Thus, the heat generated by internal resistance can be dissipated in the thermal mass of the fuel cell. This heat is then
25 passively removed by a continuous flow of air that circulates through the fuel cell stack. The fuel cell can be cooled to ambient temperature during this quiescent period and also recharged with oxygen from the flowing air stream.

There are three basic discharge modes for the pulse fuel cell, a constant current discharge, constant voltage discharge and a constant load discharge. Figure 7 shows a
30 notational constant current discharge where the voltage decays from open circuit to the maximum power at 0.50 volts. As the voltage drops from open circuit to 0.50 volts, oxygen adsorbed on the cathode generates the OH⁻ for the electrochemical reaction. The

discharge to 0.50 volts will utilize about 1/2 the oxygen stored on the electrode. The discharge cycle is followed by the quiescent period where the cathode is recharged with oxygen.

5 Constant voltage discharge will result in a time-varying current output that decays to 0 once the oxygen adsorbed in the cathode is depleted. Constant load or constant resistance discharge is characterized by the voltage and current both displaying exponential decrease as a function of time.

EXAMPLES

10 A series of laboratory tests were performed by Procyon to verify the peak power fuel cell concept. These tests were performed on numerous 1/2 cell cathodes including silver, graphite, activated carbon, and a composite electrode containing various portions of graphite and activated carbon. Anode 1/2 cell testing was performed on several high performance carbon anodes with platinum catalyst loading of $4\text{mg}/\text{cm}^2$. These anodes were
15 tested at ambient temperature and elevated temperatures of 65 degrees C, typical of conventional fuel cell operating temperature. Finally, complete single cell tests were performed on the anodes and cathodes together.

20

Example 1 -- Comparison of Silver with Carbon/Graphite Half-cell Tests in Oxygen

Figure 8 shows the data with oxygen at two different sweep rates: a 30 second sweep rate (0.25 A/sec) representing steady state operation, and the fast sweep rate of 0.3
25 second (25 A/sec) representing pulse operation. At the fast sweep rate of 0.3 seconds, the polarization curves are identical for both the silver and the inventive Carbon/Graphite electrodes. The steady state, or slow sweep rate data show the significantly higher performance of the silver electrode on air. Computations integrating the current between the steady state 25 second sweep rate and the fast sweep rate of 0.25 seconds show that 8
30 coulombs/ cm^2 of current is stored on the surface of the carbon electrode, and this is independent of whether air or oxygen is the reactant. The oxygen adsorption is calculated to be 0.0013 grams of oxygen/ cm^2 .

Example 2 -- Carbon/Graphite Electrode 1/2 Cell Tests on Air and Oxygen

A desirable variation of the carbon/graphite electrodes is a mixture of 60% activated carbon, 20% graphite, with the balance as inert binder. This mixture is rolled onto a current collector screen of nickel or silver weighing 100 to 200 mg/cm². This implementation of the pulse fuel cell uses no catalyst and operates best at ambient temperature where oxygen adsorption is maximized.

The same test procedure was used to evaluate graphite electrodes operating on air verses operating on pure oxygen. The results of these tests at the same sweep rates are shown in Figure 9 for air and oxygen. The performance of the carbon/graphite electrode on air at the high sweep rate of 0.25 seconds actually exceeds the performance of the silver electrode at high current densities greater than 800 mA/cm². It is important to note that no catalyst was used in the inventive Carbon/Graphite electrodes used in these tests, and the data was taken at 20 °C or ambient temperature. The data taken at slow sweep rates representing steady state performance shows that the silver electrode has significantly higher performance than the non-catalyzed graphite/carbon cathode.

Example 3 Hydrogen Electrode Performance Limit

For an alkaline system, the maximum pulse current that can be drawn is, in the limit, defined by the polarization of the hydrogen electrode. In a conventional alkaline system, the performance is limited by the losses that occur in the oxygen electrode. When operating the alkaline fuel cell in the pulse mode, the activation and concentration polarization are eliminated, and the hydrogen electrode becomes the performance limiting factor.

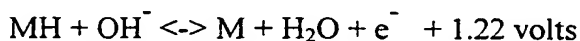
Conventional Hydrogen Electrode with Catalyzed Carbon.

The performance of a conventional catalyzed carbon hydrogen electrode in an alkaline electrolyte with a platinum loading of 4 mg Pt/cm², is shown in Figure 10. The maximum power that can be drawn from this hydrogen 1/2 cell occurs at 0.5 volts at 1 Amps/cm² at a temperature of 65°C. At ambient temperature, the current density at 0.5 volts is significantly reduced to 1/2 A/cm². Unfortunately, oxygen adsorption on the carbon cathode favors operating the fuel cell near ambient temperature. Higher platinum loading can improve the hydrogen performance at low temperatures to some degree. In order to fully optimize the performance of the pulse fuel cell, the hydrogen electrode

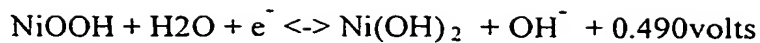
should be capable of adsorbing hydrogen, for the same reasons that oxygen adsorption on the cathode is desired. As seen from Figure 6, hydrogen adsorption on activated carbon is at least an order of magnitude lower than that of oxygen. The solution to this problem is to replace the conventional hydrogen electrode with a metal hydride electrode designed to adsorb sufficient hydrogen to satisfy the coulombic requirements of the pulse fuel cell. The metal hydride must also be electrochemically reversible, that is, hydrogen adsorption and activated hydrogen desorption are necessary. To accomplish this reversibility, an important aspect of the design is the range of metal-to-hydrogen bond strengths, which must be between 6 to 12 kcal/mole. If the bond strength is too weak hydrogen will not adsorb, and will remain as a gas. If the bond strength is too strong, the metal will be oxidized.

Improved Hydrogen Electrode Performance

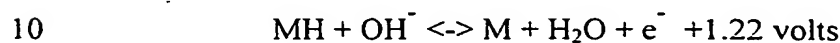
The pulse performance of the hydrogen electrode can be improved by incorporating metal hydride materials in the anode. This method will eliminate the limitations imposed by operating the hydrogen electrode in the conventional manner as described above. Substituting a commercially available metal electrode (M composed of an alloy of V, Ti, Zr, Ni, Cr, Co, or Fe or other known hydride-forming metals or alloys) is substituted for the conventional hydrogen electrode. This alloy M, is comprised of a number of other metals to optimize hydrogen adsorption, and oxidation or corrosion resistance. This is the same technology that has been developed for constructing nickel metal hydride batteries. It has been developed to a high degree, and is commercially available. This method of increasing the performance of the hydrogen electrode at low temperatures is attractive since hydrogen adsorption is favored at low temperatures, as it is in my inventive cathode, where oxygen is adsorbed on carbon. The anode reaction is for a NiM hydride battery is:



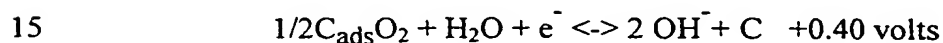
For instance, when the cathode in the metal hydride batteries is composed of nickel hydroxide $\text{Ni}(\text{OH})_2$, which is converted to nickel oxyhydroxide (NiOOH), the cathodic reaction is:



5 In my inventive pulse fuel cell, the anode reactions are identical to those of the metal hydride battery. The capacity or quantity of the metal hydride used in the anode will be an order of magnitude less than for the battery systems, because the electrode will be recharged on a time scale of the order of the pulse length. The reaction at the anode is identical to the reaction at the anode of a NiM hydride battery:



And at the cathode which uses a carbon based electrode as described in this patent, where oxygen is adsorbed on the carbon as follows:



With the above implementation, only internal resistance limits the performance of the pulse fuel cell.

20 **Example 4 Single Cell Test Results with Catalyzed Carbon Anodes and Carbon Graphite Cathodes**

Figure 11 depicts the results of experiments performed to extend the pulse width of a single carbon cathode half cell. This baseline electrode was charged by air and
25 demonstrated a constant current density output of 0.375 amps/cm² at 0.55 volts for a pulse duration of 0.70 seconds. The carbon content of this single baseline half cell was 20 mg carbon/ cm², and is typical of the construction of conventional alkaline fuel cell electrode.

A similar single carbon electrode was electrolytically recharged, and delivered a pulse duration of 1.4 seconds as seen in Figure 11.

30 The carbon content of the double electrode was more than doubled to 44 mg Carbon/ cm² by essentially placing two electrodes back to back, as depicted in Figure 14, and electrically connecting them in parallel. It can be seen that this electrode configuration using air charging of oxygen extended the pulse duration to 2.2 seconds, more than 3.0

times the baseline single electrode output of 0.70 seconds. This was achieved by increasing the carbon content by a factor of 2.2.

The longest pulse duration was produced by a double layer electrode with 28 mg carbon/ cm² that was electrolytically recharged and used the Eloflux method. The pulse duration was measured at 2.6 seconds. This electrode incorporated the Eloflux method is described in the appendix. The baseline single layer electrode without Eloflux was capable of producing a 1.4 second pulse. Here a factor of 1.65 increase in carbon loading produced a 1.85 increase in pulse duration. This clearly demonstrates the effectiveness of a flow-through electrode in enhancing the performance of the pulse fuel cell.

Figure 12 depicts a method of layering the electrodes to increase the carbon content. This method was used in producing electrodes to obtain the data of Figure 11. A standard method developed by Gaskatel of Kassel, Germany is used for fabrication these electrodes. The product of this electrode manufacturing process is a continuous, flexible homogeneous electrode tape. The thickness of the electrode is 0.35 mm by 100 mm wide, the length is continuous, and can be cut to any length desired. In order to produce electrodes for the pulse fuel cell experiments, a baseline single layer cell was produced. This baseline single electrode is representative of a conventional alkaline fuel cell performance. Additional electrodes of 2 layers and 4 layers are edge connected, electrically in parallel were produced as shown in Figure 12. Test data was collected on the single layer electrodes and the double layer electrodes and is shown in Figure 12.

Appendix 1

DEFINITIONS

Concentration Polarization

Concentration polarization results in a voltage drop associated with the mass transport of the reacting species in the fuel cell, and is greatest at high current densities. As the reactant is consumed by the electrochemical reaction, the concentration of the reactant is locally reduced near the reaction site. There are two types of concentration polarization, the first is associated with polarization of the electrolyte ions on the surface of the electrode, and the second is due to depletion of the reactant gases at the electrode. In conventional fuel cells the concentration polarization can be reduced by actively pumping the electrolyte thru the fuel cell, and by increasing reactant pressure, and temperatures. Figure 1 illustrates this concentration polarization in a PEM fuel cell. At sweep rates of 1 to 3 seconds, the concentration polarization is greatest, and diminishes as the sweep rate is reduced to 0.1 second. can essentially be eliminated at higher sweep rates. In the pulse power fuel cell the concentration polarization is negligible during the period of the pulse, and once the reactants are depleted from the surface of the active elements of the electrode, the concentration polarization becomes the predominant loss mechanism.

Activation Polarization

Activation losses result from the chemisorption of the reactant on the active surface catalyst of the electrode. This involves the breakage of molecular bonds of the reactant gases, and formation of new bonds between the activated reactant atoms and the catalyst. The activation loss shows up at the open circuit voltage of a fuel cell, which is typically 1.0 volt for a PEM fuel cell, and 1.1 volts for an alkaline fuel cell. The theoretical open circuit voltage of any hydrogen oxygen fuel cell is 1.23 volts, and the difference between the open circuit voltage and the theoretical voltage is the loss due to the activation polarization. It is customary to express the activation polarization ΔV as follows:

$$\Delta V_{\text{activation}} = RT / \alpha n F \ln(I/I_0)$$

Where R is the universal gas constant 8.123 joules/gram mole, and F is Faraday's constant 96500 coulombs/equivalent, and α is the transfer coefficient. For an alkaline system, the number of electrons transferred is $N=2$ and $\alpha = 0.75$, which results in an activation polarization of 0.20 volts at open circuit, and an additional polarization of 0.2
5 volts per decade of current density increase.

Eloflux System

In a conventional fuel cell, the gas supply flows parallel to the back side of the electrode, and the electrolyte flows between the electrodes carrying off heat and reaction
10 water. The Eloflux electrodes are constructed so that gas flows parallel to the electrode in the conventional way, as well as creating a flow of electrolyte thru the electrode. The electrolyte flow through the cell removes heat and water as well as decreasing the electrochemical losses associated with activation and concentration polarization. This process is incorporated in alkaline fuel cells produced by Gaskatel AG of Kassel,
15 Germany.

I CLAIM AS MY INVENTION:

1. A cathode in a fuel cell comprising:
 - a.) a current collector,
 - 5 b.) a shaped composition of activated carbon and a conduction medium electrically connected to said connector.
2. The cathode of claim 1 wherein the conduction medium comprises graphite.
- 10 3. The cathode of claim 2 wherein the shaped composition consists essentially of activated carbon and graphite.
4. The cathode of claim 3 wherein the shaped composition consists essentially of 30-65% activated carbon, 10-35% graphite, and 0-25% inert binder.
- 15 5. The cathode of claim 2 wherein the shaped composition is catalyst-free.
6. The cathode of claim 2 where the activated carbon is present in an amount of at least 75 mg/cm² and is capable of storing at least about 100 mg/cm² of O₂.
- 20 7. The cathode of claim 2 where the activated carbon comprises an aerogel.
8. The cathode of claim 2 wherein said cathode is conformed to allow electrolyte to flow through said cathode.
- 25 9. A pulse power fuel cell comprising:
 - a.) a cathode in a fuel cell comprising:
 - i.) a current collector,
 - ii.) a shaped composition of activated carbon and a conduction
 - 30 medium electrically connected to said connector, and
 - b.) an anode.

10. The pulse power fuel cell of claim 9 wherein the cathode conduction medium comprises graphite.

5 11. The pulse power fuel cell of claim 10 wherein the cathode shaped composition consists essentially of activated carbon and graphite.

12. The pulse power fuel cell of claim 10 wherein the cathode shaped composition consists essentially of 30-65% activated carbon, 10-35% graphite, and 0-25% inert binder.

10 13. The pulse power fuel cell of claim 9 wherein the cathode shaped composition is catalyst-free.

14. The pulse power fuel cell of claim 9 where the activated carbon is present in an amount of at least 75 mg/cm² and is capable of storing at least about 100 mg/cm² of O₂.

15 15. The pulse power fuel cell of claim 10 where the activated carbon comprises an aerogel.

20 16. The pulse power fuel cell of claim 9 wherein the fuel cell is substantially free of concentric polarization and activation polarization.

17. The pulse power fuel cell of claim 9 wherein the anode comprises an hydride-forming metal or alloy.

25 18. The pulse power fuel cell of claim 9 wherein the cathode further comprises a composition selected from the group consisting of hydroxyl-containing materials in which the hydroxyl is released by small voltage swings.

30 19. The pulse power fuel cell of claim 18 wherein said composition selected from the group consisting of Ag/AgO, Ni(OH)₂, and Pt(OH).

20. The pulse power fuel cell of claim 9 further comprising an alkaline electrolyte.

21. The pulse power fuel cell of claim 9 further comprising an acidic electrolyte.

22. The pulse power fuel cell of claim 9 further comprising a polymeric electrolyte membrane.

5

23. The pulse power fuel cell of claim 9 wherein said cathode is conformed to allow electrolyte to flow through said cathode.

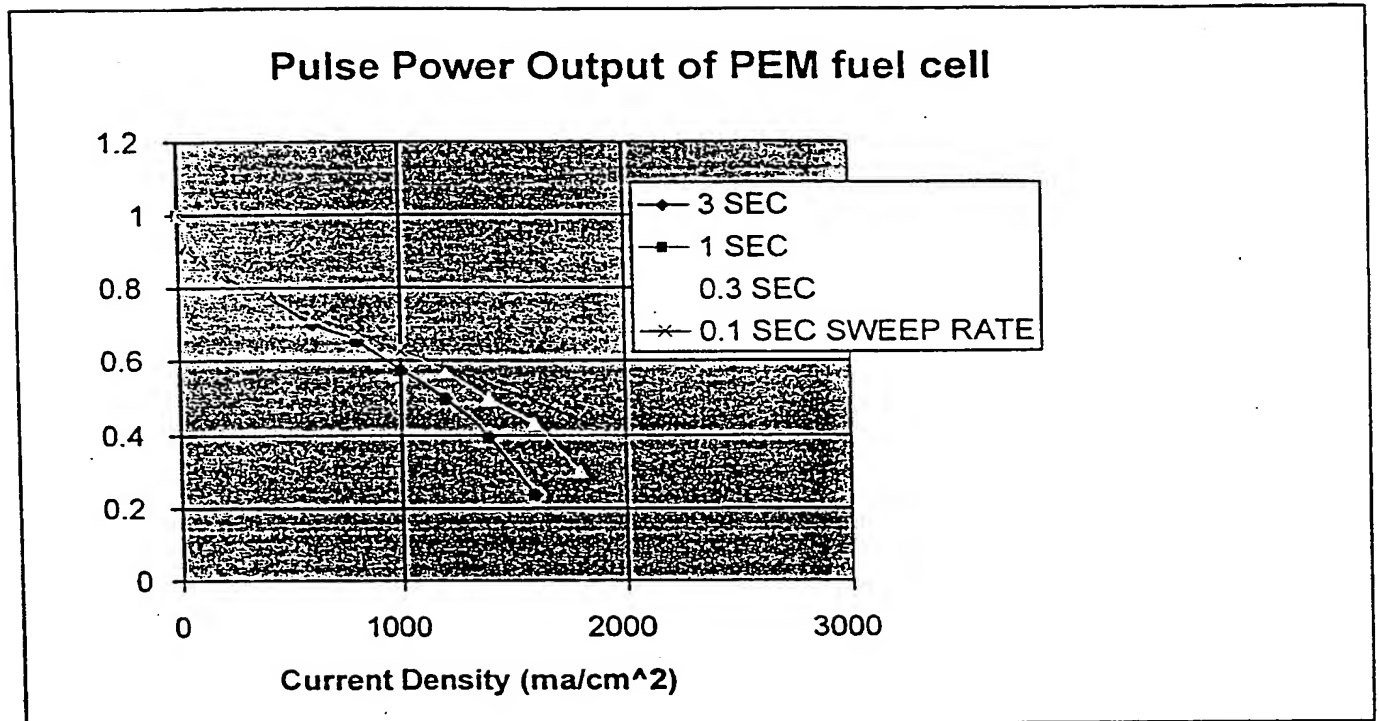


Figure 1

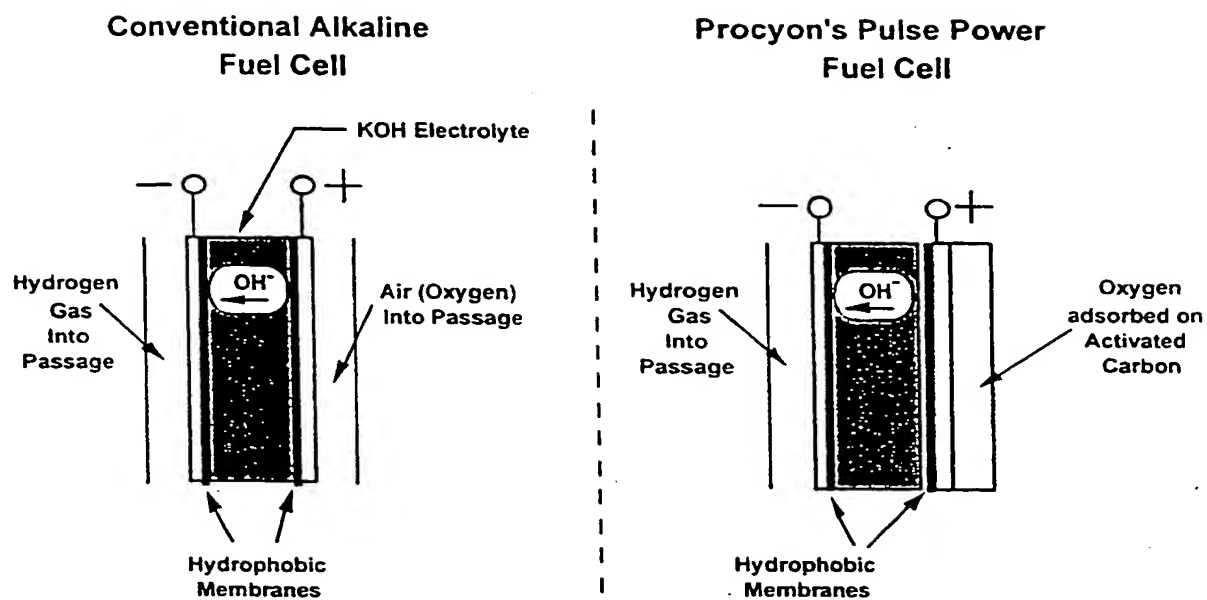
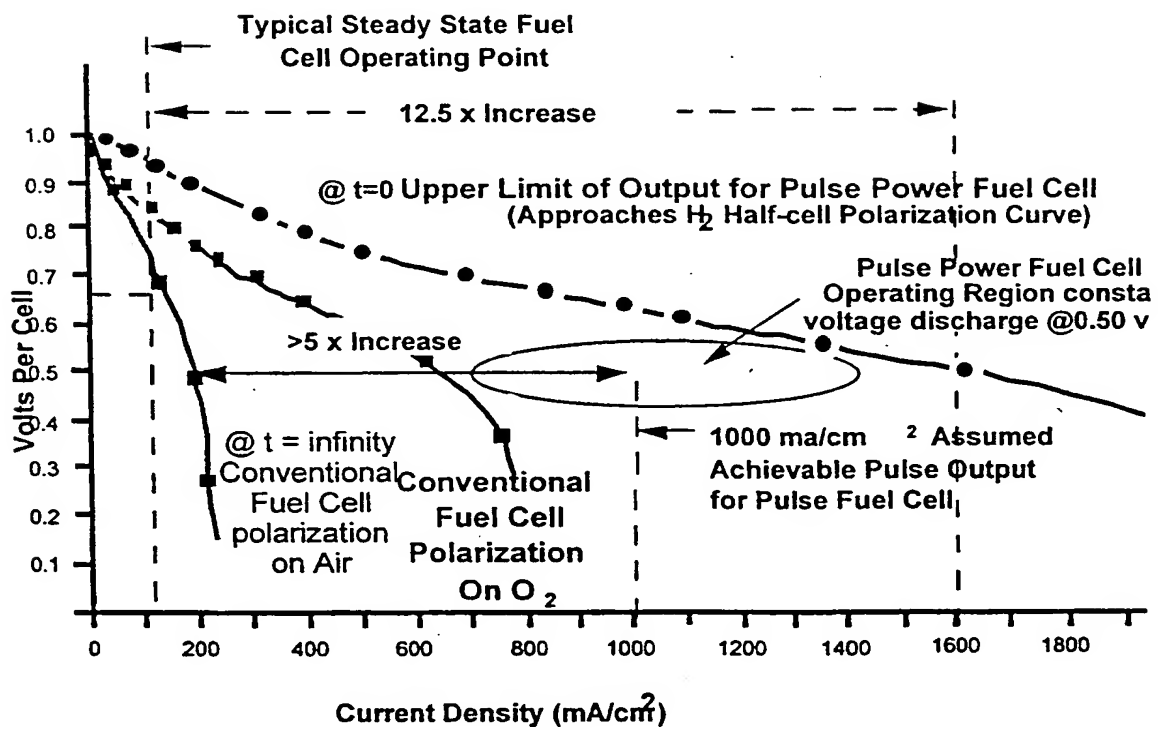


Figure 2. Procyon's Pulse Power Fuel Cell Compared to Typical Alkaline Fuel Cell

**Figure 3. Polarization Curves of Conventional Fuel Cell
Compared With a Peak Power Fuel Cell**



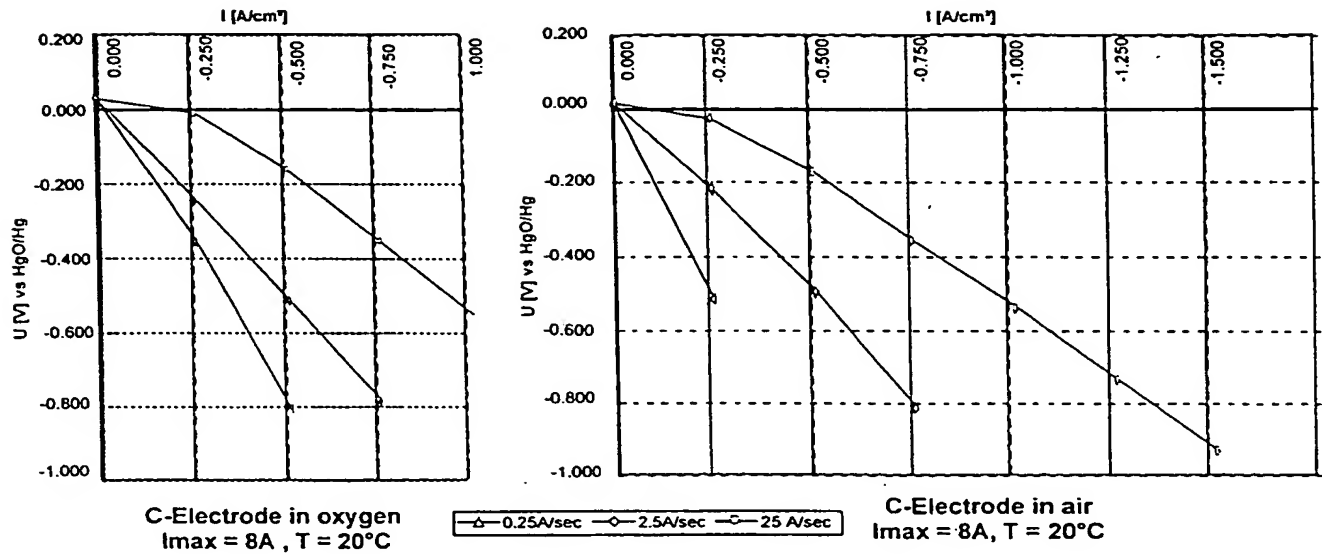


Figure 4. Polarization Curves of Conventional vs. Peak Power Fuel Cell

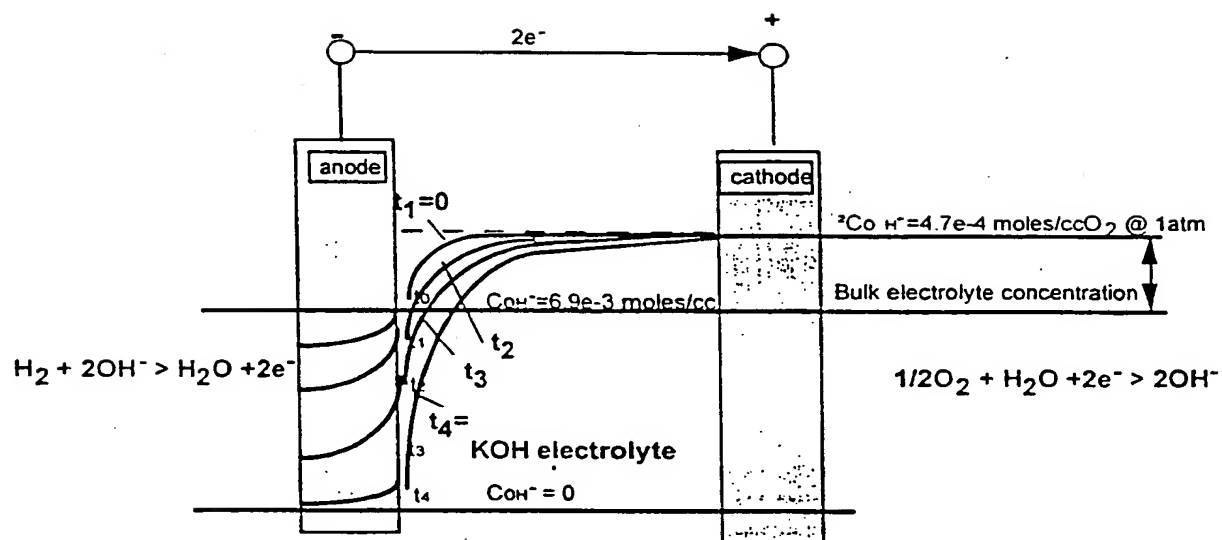


Figure 5. OH^- ion Concentration Between Electrodes as a Function of Time

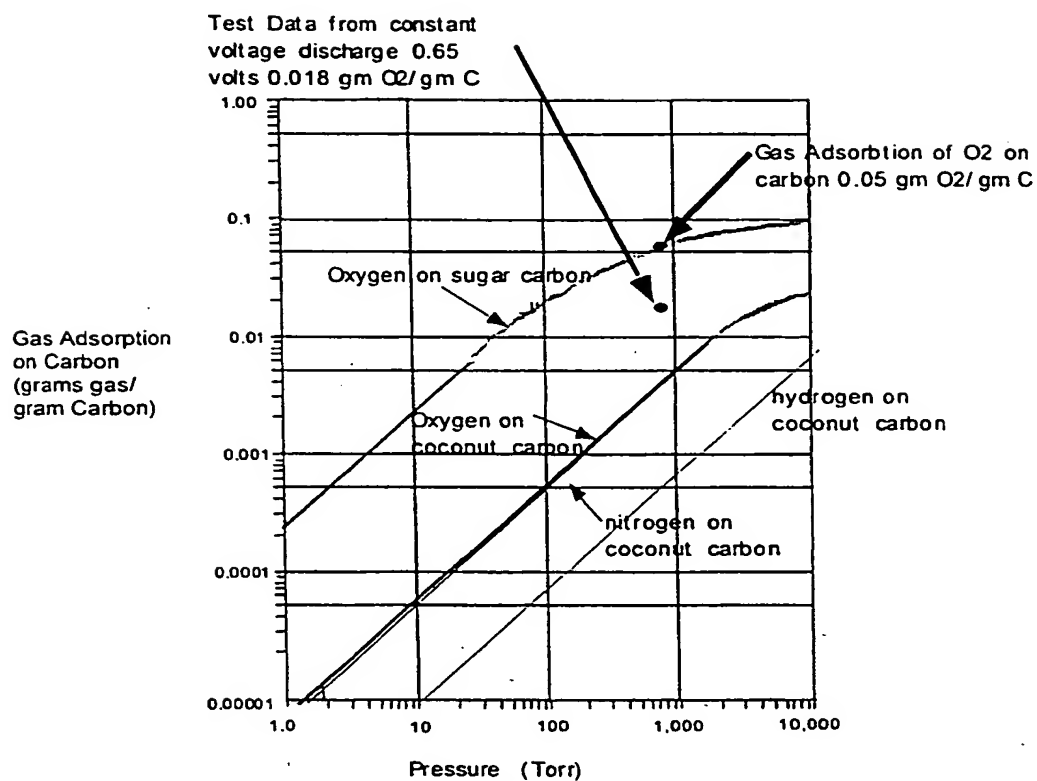


Figure 6. Oxygen Adsorbed on Activated Carbon

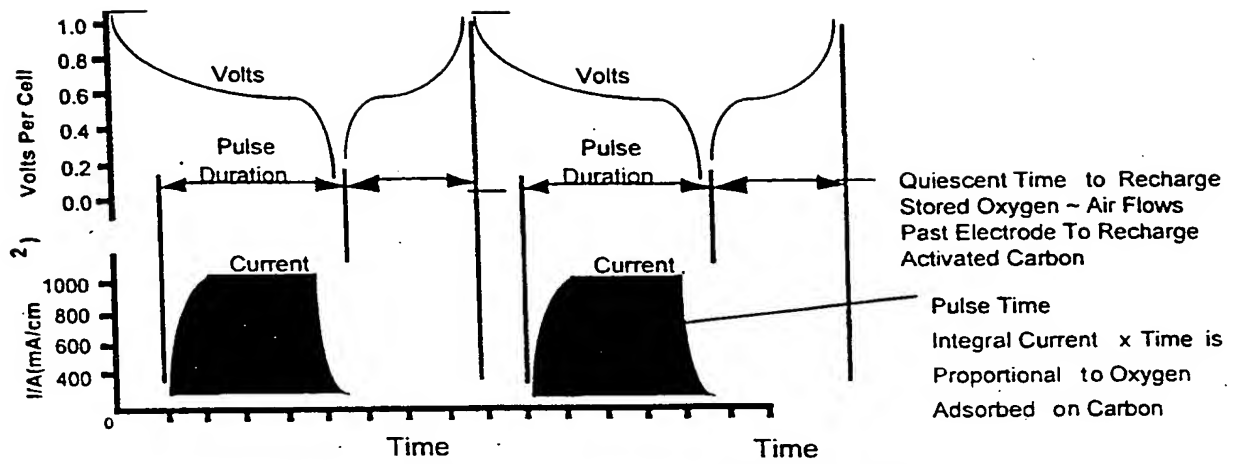


Figure 7 Constant Current Discharge Mode

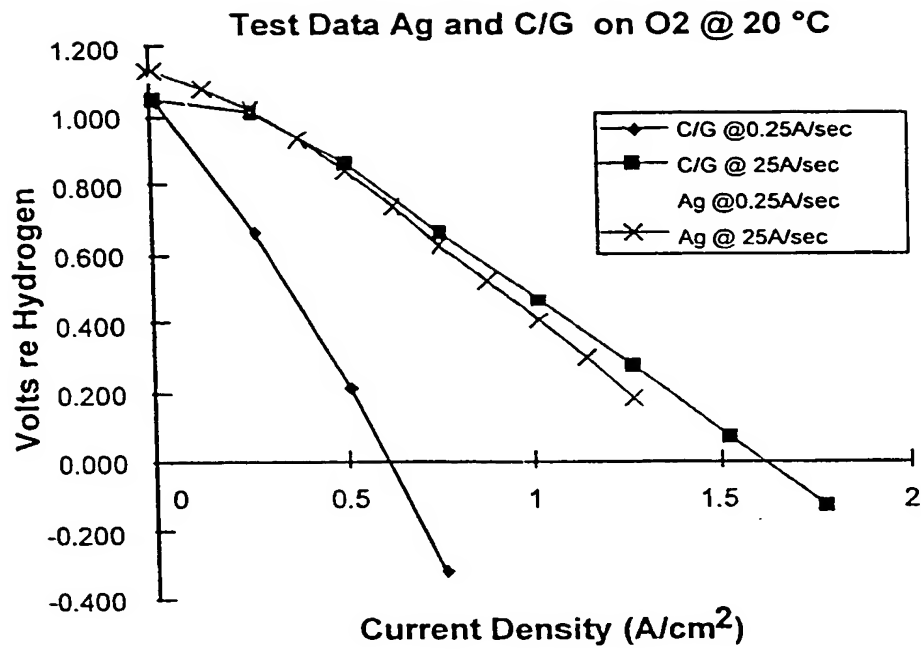


Figure 8 Performance of Silver and Carbon Cathodes with Oxygen

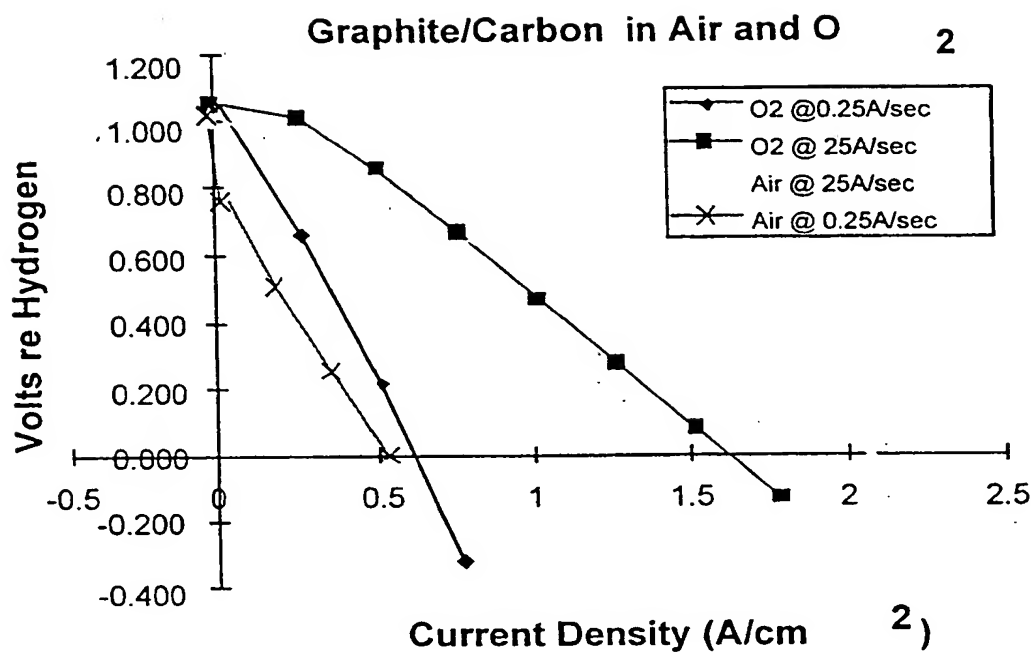
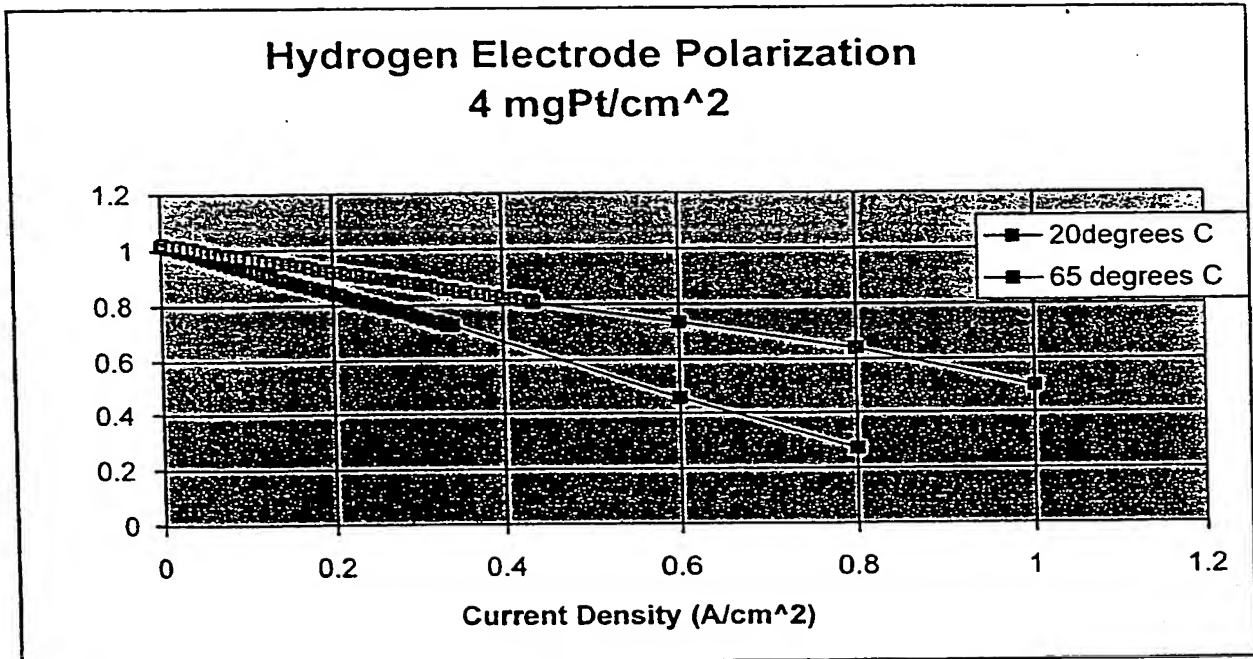


Figure 4, Carbon Electrodes Operating With Air and Oxygen

*Figure 10*

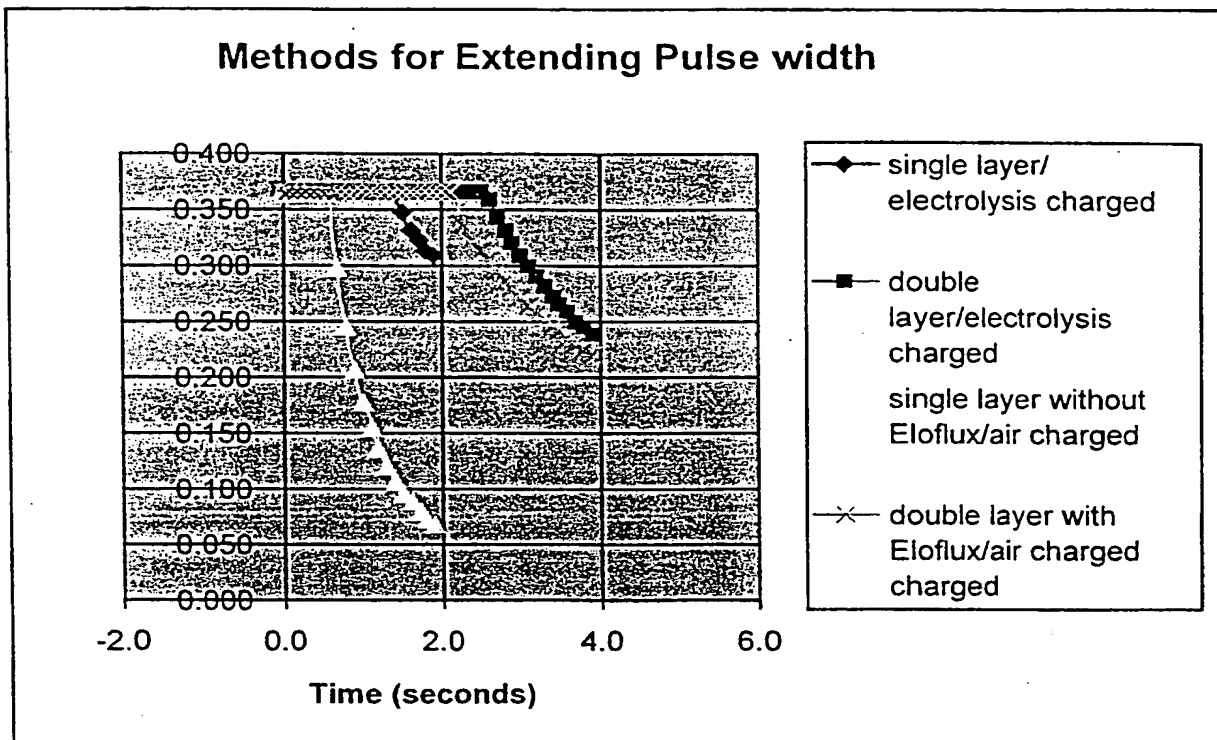
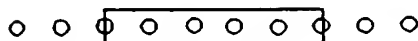


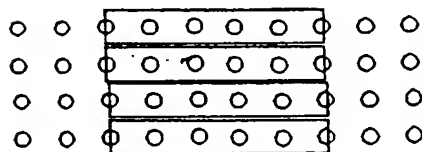
Figure 11

Screen 4cm diameter

Carbon mixture 2cm pressed onto the screen



Four electrodes (see above) pressed onto each other



Fourfold electrode (see above) connected to the electric wires in the half cell

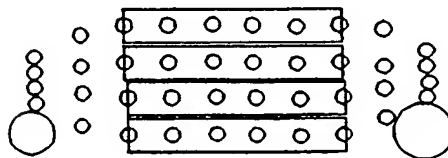


Figure 12 Configuration of multiple Electrode

INTERNATIONAL SEARCH REPORT

Intern. Application No

PCT/US 00/40268

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 H01M4/96 H01M8/08 H01M8/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

PAJ, EPO-Internal, WPI Data, CHEM ABS Data, INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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X	GB 1 153 337 A (KURT SCHWABE AND EBERHARD HOLLAX) 29 May 1969 (1969-05-29) page 1, line 11 - line 24 page 2, line 10 - line 30 column 2, line 76 - line 97 example 3.	1-4

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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

30 November 2000

Date of mailing of the international search report

07/12/2000

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Gamez, A

INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	LAKEMAN J B ET AL: "Solid polymer fuel cells for pulse power delivery" JOURNAL OF POWER SOURCES, CH, ELSEVIER SEQUOIA S.A. LAUSANNE, vol. 65, no. 1, 1 March 1997 (1997-03-01), pages 179-185, XP004059647 ISSN: 0378-7753 page 179, left-hand column, paragraph 3 -right-hand column, paragraph 1 page 180, left-hand column, paragraph 1 ----	9,22
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Intern. Application No

PCT/US 00/40268

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